

One-dimensional hyperbolic transport: positivity and admissible boundary conditions derived from the wave formulation

Antonio Brasiello¹, Silvestro Crescitelli², and Massimiliano Giona^{*3}

¹Dipartimento di Ingegneria Industriale Università degli Studi di Salerno via Giovanni Paolo II 132, 84084 Fisciano (SA), Italy

²Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale Università degli Studi di Napoli “Federico II” piazzale Tecchio 80, 80125 Napoli, Italy

³Dipartimento di Ingegneria Chimica DICMA Facoltà di Ingegneria, La Sapienza Università di Roma via Eudossiana 18, 00184, Roma, Italy

* massimiliano.giona@uniroma1.it

Abstract

We consider the one-dimensional Cattaneo equation for transport of scalar fields such as solute concentration and temperature in mass and heat transport problems, respectively. Although the Cattaneo equation admits a stochastic interpretation - at least in the one-dimensional case - negative concentration values can occur in boundary-value problems on a finite interval. This phenomenon stems from the probabilistic nature of this model: the stochastic interpretation provides constraints on the admissible boundary conditions, as can be deduced from the wave formulation here presented. Moreover, as here shown, energetic inequalities and the dissipative nature of the equation provide an alternative way to derive the same constraints on the boundary conditions derived by enforcing positivity. The analysis reported is also extended to transport problems in the presence of a biasing velocity field. Several general conclusions are drawn from this analysis that could be extended to the higher-dimensional case.

Keywords: Transport phenomena, positivity of operators, hyperbolic equations, Cattaneo equation, microscopic stochastic models.

1 Introduction

Cattaneo transport equation for scalar fields (matter, heat) stands as a valuable attempt to overcome the intrinsic problems of Fickian transport,

namely its infinite velocity of propagation [1] (throughout this article we refer as “Fickian” to the wealth of transport problems where the flux of the transported entity is proportional to the concentration gradient, such as in the Fick’s equation for mass transport, or to temperature gradient, such as in the Fourier equation for conductive heat transfer).

This is achieved by introducing an exponentially decaying memory contribution in the flux-concentration constitutive equation. The Cattaneo model finds important applications in some specific transport problems (such as the phenomenon of the second sound in superfluid helium) [2] (see also the review by Joseph and Preziosi [3, 4]), and in a variety of other, more engineering-oriented, studies [5, 6, 7]. Some authors have developed an approximate mesoscopic derivation of Cattaneo-like transport equations in the form of a generalized hyperbolic Fokker-Planck equation [8, 9], and Cattaneo model rests as a fundamental example of generalized constitutive equation in the theory of extended irreversible thermodynamics [10, 11, 12].

Its implications in theoretical physics should not be underestimated, as it represents a valuable contribution for extending transport models in a relativistic (Lorentz-invariant) way [13], and it constitutes the starting point for attempting a stochastic interpretation of the Dirac equation [14, 15].

However, its validity has been deeply questioned and criticized for space dimension greater than one, as the associated Green function attains negative values [16], so that the propagation of generic non-negative initial conditions can return negative concentration values.

In point of fact, even this observation can be pushed forward, since there exist one-dimensional problems on the finite interval for which the solution of the Cattaneo equation in the presence of non-negative initial and boundary conditions can become negative. This is shown in Section 2 by means of a simple, closed-form example. The Cattaneo equation on the interval has been studied also in [17, 18], for boundary conditions that do not present problems as it regards positivity.

This result may seem in contradiction with the stochastic derivation of the Cattaneo model starting from a simple stochastic equation driven by dichotomous Poisson noise, developed earlier by S. Goldstein [19] and subsequently re-elaborated by M. Kac in a simple but seminal contribution [20]. This contradiction is however purely apparent and can be resolved by formulating the transport problem in the wave-formalism envisaged by Kac [20] and subsequently elaborated by several others authors [18, 21, 22, 23], just to cite some relevant contributions in the field. The wave analysis indicates unambiguously that not all of the boundary conditions, that are commonly applied in a Fickian framework, are physically admissible in hyperbolic transport schemes. More precisely, the wave-like nature of the transport models with memory dictates bounds and constraints on the admissible boundary conditions preserving positivity. This is shown in Sections 3 and 4 dealing with the desorption experiment addressed in Section 2. The

same constraints derived from positivity emerge from the analysis of the L^2 -norms, i.e., from energetic conditions, once dissipation is enforced (see Section 5).

In this paper the analysis is also extended to one-dimensional problems in the presence of a deterministic biasing velocity field, deriving constraints on boundary conditions in the classical problem of boundary-layer polarization characterizing transport across permeable or perm-selective membranes (see Section 6). Some general conclusions, oriented towards a rigorous formulation of hyperbolic transport models, are addressed in the concluding Section 7.

2 The Cattaneo equation on the unit interval

Consider a transport problem for a conserved scalar field $u(x, t)$ on the spatial interval $x \in [0, L]$ and time t , in the absence of source terms. In this case, the balance equation is obviously given by

$$\frac{\partial u(x, t)}{\partial t} = -\frac{\partial J(x, t)}{\partial x}, \quad (1)$$

and assume that the flux $J(x, t)$ is related to $u(x, t)$ via a constitutive equation of Cattaneo type

$$J(x, t) = -k \frac{\partial u(x, t)}{\partial x} - \tau_c \frac{\partial J(x, t)}{\partial t}, \quad (2)$$

where k is the “Fickian” diffusivity (conductivity) and τ_c the characteristic memory time. By substituting eq. (2) into eq. (1) we obtain

$$\frac{\partial u(x, t)}{\partial t} + \tau_c \frac{\partial^2 u(x, t)}{\partial t^2} = k \frac{\partial^2 u(x, t)}{\partial x^2}, \quad (3)$$

namely the Cattaneo equation.

As $\tau_c \rightarrow 0$, the Cattaneo equation degenerates into a diffusion equation, while as $\tau_c \rightarrow \infty$, keeping $k/\tau_c = \text{constant}$, it reduces to a one-dimensional wave equation. Both these two limit conditions, i.e., the parabolic limit and the pure dissipation-free wave equation, bring physical contradiction inside, namely the infinite velocity of propagation characterizing Fickian transport on one hand, and the occurrence of pure wave-like propagation without dissipation on the other hand, as mentioned by Joseph and Preziosi [3, 4].

As initial condition at $t = 0$ let

$$u(x, 0) = u_{\text{in}} = \text{constant}, \quad \left. \frac{\partial u(x, t)}{\partial t} \right|_{t=0} = 0. \quad (4)$$

Introducing the nondimensional variables $y = x/L$, $\theta = tk/L^2$, $\alpha = \tau_c k/L^2$, $\phi = u/u_{\text{in}}$, the balance equation for $\phi(y, \theta)$ reads

$$\frac{\partial \phi}{\partial \theta} + \alpha \frac{\partial^2 \phi}{\partial \theta^2} = \frac{\partial^2 \phi}{\partial y^2}, \quad (5)$$

and $\phi|_{\theta} = 1$, $\partial\phi/\partial\theta|_{\theta=0} = 0$.

Let us consider a desorption experiment, meaning that $u(x, t)$ can be viewed, for instance, as the concentration of a solute inside some solid matrix where it diffuses. Suppose that the boundary at $x = 0$, ($y = 0$), is perfectly impermeable to transport so that $J|_{x=0} = 0$ for all $t > 0$. Conversely, at $x = L$, ($y = 1$), solute diffuses out of the solid matrix onto an external reservoir that is much larger than the sample matrix and perfectly mixed so that the concentration at $y = 1$ can be assumed vanishingly small, i.e.,

$$\phi(1, t) = 0 . \quad (6)$$

It follows from the constitutive equation, that the zero-flux boundary condition at $y = 0$ reduces to

$$\left. \frac{\partial\phi(y, \theta)}{\partial y} \right|_{y=0} = 0 . \quad (7)$$

From eqs. (6)-(7) the concentration field $\phi(y, \theta)$ can be expanded in Fourier series

$$\phi(y, \theta) = \sum_{n=1}^{\infty} \phi_n(\theta) \cos(\nu_n y) , \quad \nu_n = \frac{(2n-1)\pi}{2} . \quad (8)$$

The Fourier coefficients $\phi_n(\theta)$ satisfy the equation

$$\alpha \ddot{\phi}_n(\theta) + \dot{\phi}_n(\theta) + \nu_n^2 \phi_n(\theta) = 0 , \quad (9)$$

where $\dot{\phi}_n$ indicates the derivative with respect to θ and $\ddot{\phi}_n$ the second derivative, equipped with the initial condition $\phi_n(0) = 2(-1)^{n-1}/\nu_n = \phi_{n,0}$, $n = 1, 2, \dots$. The characteristic equation associated with eq. (9) is readily $\lambda^2 + \lambda/\alpha + \nu_n^2/\alpha = 0$, and it admits the solutions

$$\lambda = -\frac{1}{2\alpha} \pm \frac{\sqrt{\Delta_n}}{2\alpha} , \quad \Delta_n = 1 - 4\nu_n^2\alpha . \quad (10)$$

Depending on the sign of the discriminants Δ_n two cases occur: (i) for $n \leq n^*$, where $\Delta_{n^*} > 0$ and $\Delta_{n^*+1} < 0$, the roots of the characteristic equation are real: $\lambda_{1,n} = -1/2\alpha + \sqrt{\Delta_n}/2\alpha$, $\lambda_{2,n} = -1/2\alpha - \sqrt{\Delta_n}/2\alpha$. Therefore, for $n \leq n^*$,

$$\phi_n(\theta) = A_n e^{\lambda_{1,n}\theta} + B_n e^{\lambda_{2,n}\theta} . \quad (11)$$

Enforcing the initial conditions one gets

$$A_n = \frac{\lambda_{2,n} \phi_{n,0}}{\lambda_{2,n} - \lambda_{1,n}} , \quad B_n = -\frac{\lambda_{1,n}}{\lambda_{2,n}} A_n . \quad (12)$$

For $n > n^*$, the roots of the characteristic equation are complex conjugate: $\lambda_{1,2} = -1/2\alpha \pm i\omega_n$, $i = \sqrt{-1}$, $\omega_n = \sqrt{-\Delta_n}/2\alpha$, and

$$\phi_n(\theta) = e^{-\theta/2\alpha} [C_n \cos(\omega_n \theta) + D_n \sin(\omega_n \theta)] , \quad (13)$$

where:

$$C_n = \phi_{n,0} , \quad D_n = \frac{C_n}{2\alpha \omega_n} . \quad (14)$$

To sum up, the solution of the above transport problem involving the Cattaneo equation can be expressed as

$$\begin{aligned} \phi(y, \theta) &= \sum_{n=1}^{n^*} \left[A_n e^{\lambda_{1,n}\theta} + B_n e^{\lambda_{2,n}\theta} \right] \cos(\nu_n y) \\ &= \sum_{n=n^*+1}^{\infty} [C_n \cos(\omega_n \theta) + D_n \sin(\omega_n \theta)] e^{-\theta/2\alpha} \cos(\nu_n y) . \end{aligned} \quad (15)$$

Figure 1 depicts the concentration profiles for different values of θ along the nondimensional spatial coordinate y , for $\alpha = 0.2$ (panel a) and for $\alpha = 0.5$ (panel b). In both cases, it can be neatly observed that the concentration attains negative values.

The unphysical inconsistency of the transport model is even more evident by considering the relative released fraction

$$\frac{M_d(\theta)}{M_\infty} = 1 - \int_0^1 \phi(y, \theta) dy . \quad (16)$$

In the case under examination, since the time derivative of the flux at $y = 1$ is identically vanishing for $\theta > 0$, eq. (16) can be written in the equivalent form

$$\frac{M_d(\theta)}{M_\infty} = - \int_0^\theta \left. \frac{\partial \phi(y, \theta')}{\partial y} \right|_{y=1} d\theta' , \quad (17)$$

similarly to the pure Fickian case. The quantity $M_d(\theta)/M_\infty$ represents the ratio of solute released up to time θ to the overall solute quantity initially present within the matrix. Since there is no source, physics dictates that this quantity should be bounded by 1, i.e., $M_d(\theta)/M_\infty \leq 1$, $\lim_{\theta \rightarrow \infty} M_d(\theta)/M_\infty = 1$.

Figure 2 depicts the time evolution of $M_d(\theta)/M_\infty$ for several values of the dimensionless number α , showing the occurrence of an overshoot of this quantity reaching unphysical values greater than 1. This phenomenon is more pronounced as α increases i.e., as the influence of the memory term in the constitutive equation becomes more significant.

It should be observed that overshoot dynamics in sorption experiments have been observed in transport across polymeric matrices where the viscoelastic response of the material is responsible for the occurrence of memory effect in mass transport (see [24] and reference therein, and particularly figure 1 in this article based on closed-form solution of the transmission-line equation [25]). However, while in sorption experiments an overshoot in the overall sorption curve does not violate in principle any fundamental law

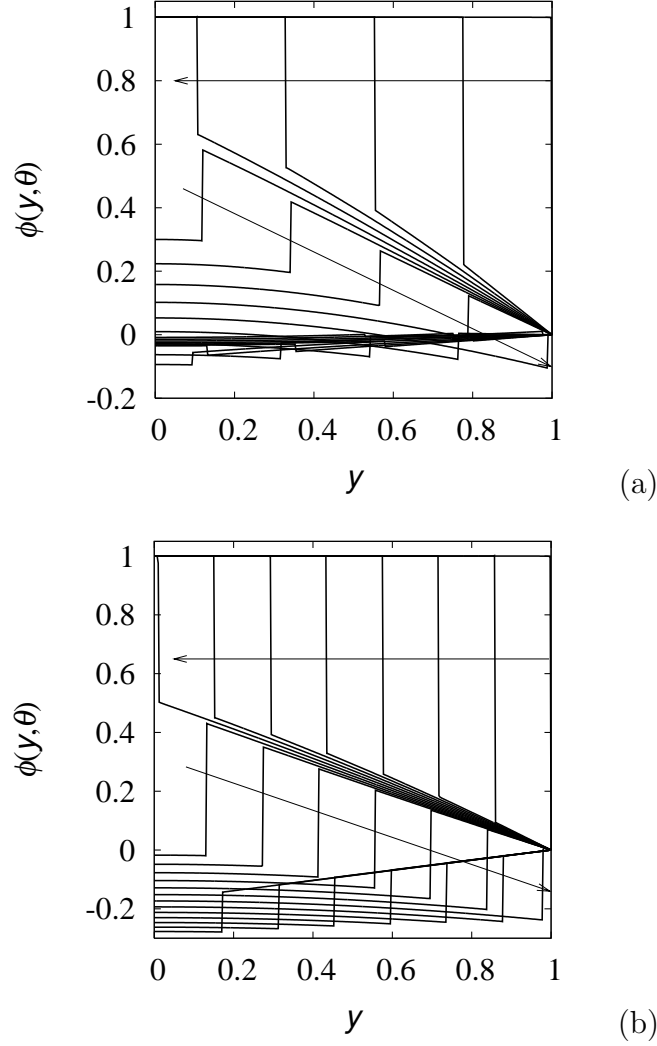


Figure 1: Concentration profiles $\phi(y, \theta)$ vs y for increasing values of $\theta = n\Delta\theta$, $n = 1, 2, \dots$, $\Delta\theta = 0.1$ (in the direction of the arrows). Data are obtained from eq. (15) truncating the series up to $N = 40000$ modes. Panel (a) refers to $\alpha = 0.2$, panel (b) to $\alpha = 0.5$.

of nature, the occurrence of $M_d(\theta)/M_\infty$ greater than 1 in the desorption experiment discussed in this Section is physically inconsistent, since it is the macroscopic manifestation of the occurrence of negative concentration values.

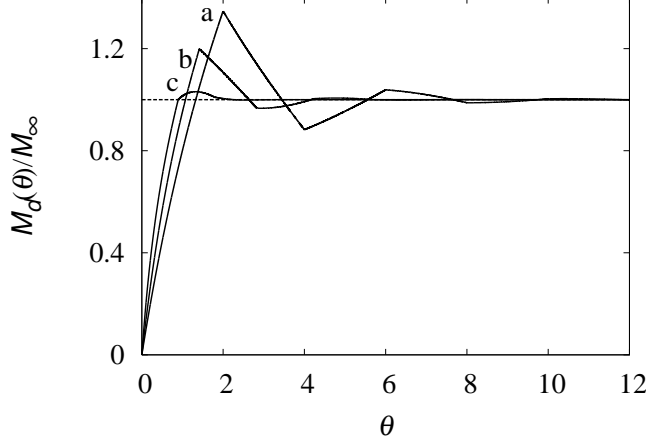


Figure 2: $M_d(\theta)/M_\infty$ vs θ for different values of α . Line (a): $\alpha = 1$, line (b): $\alpha = 0.5$, line (c): $\alpha = 0.2$.

3 Stochastic dynamics, wave formulation and boundary-condition constraints

The paradoxes of negative concentration values and of a released fraction greater than one at intermediate timescales provide an interesting example of the role played by the boundary conditions, in that the choice of admissible boundary conditions is entirely entailed in the field equations, or better to say in the present case, in its underlying microscopic stochastic formulation. Different classes of boundary conditions for the Cattaneo equation defined on an interval have been addressed in [17, 26, 27]. The focus of the present analysis is however slightly different with respect to these papers, as it is strictly oriented on the consistency of the boundary conditions in order to preserve the positivity of the solutions.

The result obtained in the previous Section could sound as a paradox, by considering that the Cattaneo equation on the one-dimensional line admits a probabilistic interpretation in terms of a stochastic process driven by dichotomous Poisson noise [20, 18, 22, 23].

Consider the stochastic equation

$$dx(t) = c(-1)^{\chi(t)} dt, \quad (18)$$

where $c > 0$ is a reference velocity, $\chi(t)$ an ordinary Poisson process characterized by the transition rate constant $a > 0$ [20]. Let $X(t)$ be the stochastic process associated with eq. (18) and let

$$\begin{aligned} p^+(x, t) dx &= \text{Prob}\{X(t) \in (x, x + dx), (-1)^{\chi(t)} = 1\} \\ p^-(x, t) dx &= \text{Prob}\{X(t) \in (x, x + dx), (-1)^{\chi(t)} = -1\}, \end{aligned} \quad (19)$$

the probabilities of the occurrence of a value of $X(t)$ in the interval $(x, x+dx)$, and of the stochastic dichotomic perturbation $(-1)^{\chi(t)} = \pm 1$, respectively, at time t . The two partial probability densities $p^\pm(x, t)$ specify completely the statistical characterization of the process (18) [20]. From eqs. (18)-(19), it follows that the evolution equations for the two quantities $p^\pm(x, t)$ read

$$\begin{aligned}\frac{\partial p^+(x, t)}{\partial t} &= -c \frac{\partial p^+(x, t)}{\partial x} - ap^+(x, t) + ap^-(x, t) \\ \frac{\partial p^-(x, t)}{\partial t} &= c \frac{\partial p^-(x, t)}{\partial x} + ap^+(x, t) - ap^-(x, t),\end{aligned}\quad (20)$$

where a is the rate constant of the Poisson process $\chi(t)$. By defining the two quantities,

$$p(x, t) = p^+(x, t) + p^-(x, t), \quad J(x, t) = c[p^+(x, t) - p^-(x, t)] \quad (21)$$

it follows that $p(x, t)$ satisfies the conservation equation

$$\frac{\partial p(x, t)}{\partial t} = -\frac{\partial J(x, t)}{\partial x}, \quad (22)$$

while $J(x, t)$ fulfills the relation

$$\frac{1}{2a} \frac{\partial J(x, t)}{\partial t} + J(x, t) = -\frac{c^2}{2a} \frac{\partial p(x, t)}{\partial x}. \quad (23)$$

The two scalar fields $p(x, t)$ and $J(x, t)$ correspond respectively to the overall probability density function and its flux associated with the stochastic dynamics (18), and eqs. (22)-(23) return the Cattaneo transport equation by identifying $\tau_c = 1/2a$ and $k = c^2/2a$. It follows from the definition, that the quantity $p(x, t)$ should be strictly positive due to its probabilistic interpretation. Let us refer to $p^+(x, t)$ and $p^-(x, t)$ as the partial probability densities (or waves), and to the equations (20) as the *partial probability model* (acronym PPM).

Starting from the decomposition into partial probabilities, it is possible to develop a theory for the boundary conditions. The partial probabilities are the fundamental physical quantities in a transport process on the line driven by dichotomous noise, while $p(x, t)$ and $J(x, t)$ should be viewed as derived quantities. Consequently, the boundary conditions should be properly expressed in terms of $p^+(x, t)$ and $p^-(x, t)$ to ensure positivity. This concept can be expressed in an alternative way: physically admissible boundary conditions for the hyperbolic transport problem on the interval $[0, L]$ are those that preserve the positivity of the partial waves $p^+(x, t)$, $p^-(x, t)$, starting from non-negative (initial and boundary) values of these two quantities. The application of this approach to the setting of the boundary conditions explains the apparent paradox addressed in Section 2.

Consider the boundary conditions for the problem addressed in Section 2, namely $J|_{x=0} = 0$ and eq. (6), i.e., in the present notation $p(1, t) = 0$. The impermeability condition at $x = 0$ implies for the partial probabilities

$$p^+(x, t)|_{x=0} = p^-(x, t)|_{x=0} , \quad (24)$$

which is a fully admissible boundary condition. Eq. (24) implies that the forward propagating wave (p^+) equals the incoming regressive wave p^- at $x = 0$. Next, consider the other boundary condition eq. (6). It means that $[p^+(x, t) + p^-(x, t)]|_{x=L} = 0$, i.e.,

$$p^-(x, t)|_{x=L} = -p^+(x, t)|_{x=L} , \quad (25)$$

implying that, if the forward probability is positive at $x = L$, the “reflected” backward wave should attain negative values. Clearly, it is not an admissible boundary condition, the consequence of which is the occurrence of negative values for $p^+(x, t) + p^-(x, t)$.

In a similar way, it is possible to address other kinds of boundary conditions. A paradigmatic example is given by radiative boundary conditions arising e.g. in interphase transport across a boundary layer. Below, we analyze this case. The prototype of homogeneous radiative boundary conditions is given by

$$J(x, t)|_{x=L} = h p(x, t)|_{x=L} , \quad (26)$$

where h (possessing the physical dimension of a velocity m/s) is the mass-transfer coefficient across a boundary layer at $x = L$. Substituting the expressions for p and J in terms of partial probabilities, eq. (26) becomes

$$p^-(x, t)|_{x=L} = \frac{c-h}{c+h} p^+(x, t)|_{x=L} . \quad (27)$$

The latter expression indicates that positivity is preserved provided that the condition

$$c - h \geq 0 \quad (28)$$

is fulfilled. This condition can be expressed in terms of the physical parameters k and τ_c resulting

$$De_h = \frac{h^2 \tau_c}{k} \leq 1 . \quad (29)$$

where the dimensionless group De_h can be referred to as the *radiative Deborah number*. The radiative Deborah number is the ratio of the characteristic “recombination time” τ_c (the intrinsic memory timescale) to the diffusion (conduction) time associated with the transfer across the boundary layer $t_{bl} = \delta^2/k$, where δ is the boundary-layer width. Since the boundary-layer width δ is implicitly defined by the relation $h = k/\delta$, it follows that $t_{bl} = k/h^2$.

Eq. (29) indicates that for the transport problem under consideration, radiative boundary conditions are admissible (i.e. they preserve positivity) provided that the Deborah number De_h is less than or at most equal to 1.

4 Revisiting the boundary conditions

In the previous Section we have derived the more general form of linear admissible boundary condition in the framework of the one-dimensional PPM:

$$p^-(y, \theta)|_{y=1} = \gamma p^+(y, \theta)|_{y=1} , \quad (30)$$

where $\gamma \in [0, 1]$ is a dimensionless parameter depending solely on the Deborah number De_h . Enforcing eq. (30) at $y = 1$, and the zero-flux boundary conditions at $y = 0$, it is possible to obtain physically consistent results regarding desorption kinetics in hyperbolic transport.

Figure 3 depicts the overall concentration profile $\phi(y, \theta) = p(y, \theta) = p^+(y, \theta) + p^-(y, \theta)$, solution of the dimensionless PPM, equipped with the boundary condition (30), $\gamma = 0$ at $y = 1$ for two different values of the dimensionless parameter α . The initial conditions are $p^+(y, 0) = p^-(y, 0) = 1/2$ uniformly in $y \in (0, 1)$. As expected from the analysis of admissible boundary conditions developed in Section 3, the occurrence of negative concentration values is “cured” by eq. (30).

The overall desorption kinetics, expressed by the relative release fraction $M_d(\theta)/M_\infty$, is depicted in figure 4. Panel (a) refers to $\gamma = 0$ for different values of α , while panel (b) depicts the influence of the boundary parameter γ . As expected, the release kinetics becomes progressively slower as γ approaches 1, since $\gamma = 1$ corresponds to the impermeability (zero-flux) condition. The data depicted in figure 4 indicate that the unphysical overshoot of $M_d(\theta)/M_\infty$ up to values greater than 1 cannot occur if proper boundary conditions are enforced, accounting for the wave-like propagation of the stochastic perturbation.

With reference to the desorption experiment analyzed throughout this work, let

$$p(y, \theta) = 1 + u(y, \theta) . \quad (31)$$

Since $0 \leq p(y, \theta) \leq 1$, it follows that $-1 \leq u(y, \theta) \leq 0$. In terms of partial waves this implies

$$p^+(y, \theta) = \frac{1}{2} + u^+(y, \theta) , \quad p^-(y, \theta) = \frac{1}{2} + u^-(y, \theta) . \quad (32)$$

The balance equations for the partial waves $u^\pm(y, \theta)$ are identical to eqs. (20) replacing p^\pm with u^\pm , t with θ and x with y . The initial conditions are $u^+(y, 0) = u^-(y, 0) = 0$, and impermeability at $y = 0$ is just $u^+(0, \theta) = u^-(0, \theta)$.

More interesting is the boundary condition at $y = 1$. From eqs. (30) and (32) one obtains

$$u^-(y, \theta)|_{y=1} = \gamma u^+(y, \theta)|_{y=1} - \frac{1-\gamma}{2} . \quad (33)$$

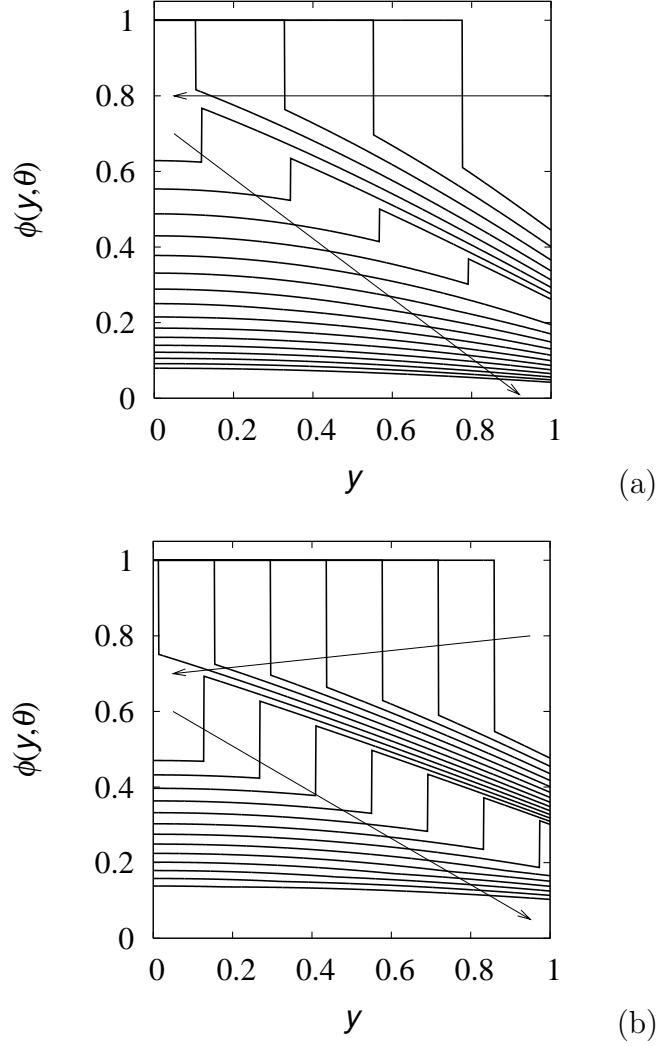


Figure 3: Concentration profiles $\phi(y, \theta)$ vs y solutions of the PPM with the boundary condition (30) at $y = 1$, $\gamma = 0$ for increasing values of $\theta = n\Delta\theta$, $n = 1, 2, \dots$, $\Delta\theta = 0.1$ (in the direction of the arrows). Panel (a) refers to $\alpha = 0.2$, panel (b) to $\alpha = 0.5$.

This indicates that the desorption kinetics can be solved by considering the complementary partial waves $u^\pm(y, \theta)$ equipped at the transfer boundary ($y = 1$) with the condition (33). This is a non-homogeneous expression containing a strictly negative source term $-(1 - \gamma)/2 < 0$, since $\gamma \in (0, 1)$.

This elementary observation suggests a way to handle boundary conditions in a sorption experiment, which is the complementary problem to the desorption kinetics analyzed so far. In a sorption experiment, solute diffuses

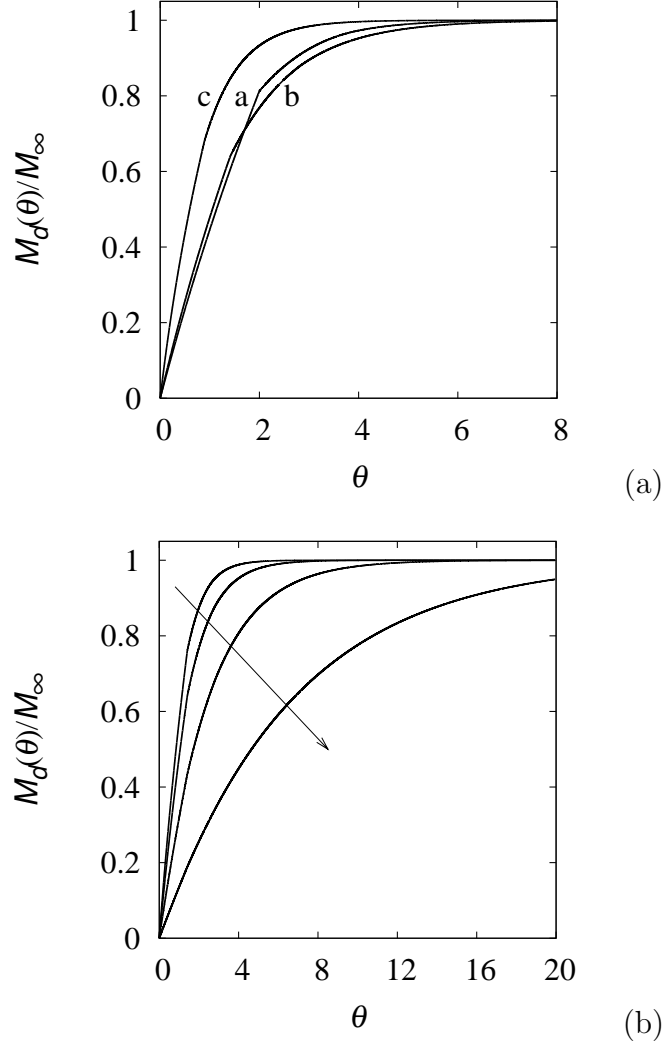


Figure 4: $M_d(\theta)/M_\infty$ vs θ obtained from the PPM. Panel (a): $\gamma = 0$, for different values of α . Line (a): $\alpha = 1$, line (b): $\alpha = 0.5$, line (c): $\alpha = 0.2$. Panel (b) $\alpha = 0.5$, for different values of γ . The arrow indicates increasing values of $\gamma = 0, 0.2, 0.5, 0.8$.

within the solid matrix from $y = 1$, where it is in contact with a perfect reservoir at constant unit dimensionless concentration. Assume a vanishing initial condition in $y \in (0, 1)$. The boundary condition at the transfer interface $y = 1$, that is complementary to the desorption condition (30) is simply

$$u^-(y, \theta)|_{y=1} = \gamma u^+(y, \theta)|_{y=1} + \frac{1-\gamma}{2}. \quad (34)$$

It corresponds to a change of sign in the source term entering the desorption conditions (33). Initial conditions are $u^+|_{\theta=0} = u^-|_{\theta=0} = 0$, and at $y = 0$ is enforced.

In a sorption experiment, the overall uptake kinetics is expressed by the sorption curve

$$\frac{M_s(\theta)}{M_\infty} = \int_0^1 [u^+(y, \theta) + u^-(y, \theta)] dy . \quad (35)$$

Figure 5 (panel a) depicts the concentration profiles for $\tau_c = 0.5$, $\gamma = 0.6$ at different time instants sampled at $\Delta\theta = 0.1$. Panel (b) of the same figure compares the resulting sorption curves (solid lines) obtained using eq. (35) and the corresponding desorption curve associated with eq. (30). As expected, sorption and desorption curves at the same values of τ_c and γ coincide, confirming that the nonhomogeneous boundary conditions (34) represents in sorption experiments the analogue of the desorption boundary conditions (30).

It is important to stress once again that this complementarity derives from the intrinsic decomposition of the concentration field into partial waves. This decomposition, with all of its implications, is the essence of hyperbolic transport. Any attempt in modeling boundary conditions in sorption dynamics starting directly from the overall concentration field $u(y, \theta)$, thus neglecting the underlying decomposition in partial waves $u^\pm(y, \theta)$, leads unavoidably to physical inconsistencies. Let us clarify this statement with a simple example. In an ideal sorption experiment (starting from $u|_{\theta=0} = 0$), the simplest boundary condition corresponding to the contact with an ideal reservoir at unit concentration would be

$$u(y, \theta)|_{y=1} = 1 , \quad (36)$$

in the absence of transfer resistances. The use of this equation, or of the equivalent expression,

$$u^-(y, \theta)|_{y=1} = 1 - u^+(y, \theta)|_{y=1} \quad (37)$$

ends up with an overshoot dynamics for the sorption curve $M_s(\theta)/M_\infty$, corresponding to local concentration values greater than 1, i.e. greater than the feeding concentration at the boundary $y = 1$. This is shown in figure 6 depicting the concentration profiles (panel a), and the overall sorption curve (panel b) at $\tau_c = 0.5$.

5 Norm dynamics and dissipation

The constraints on the structure of the boundary conditions can be derived from the evolution of the integral norms, enforcing dissipation. Consider eq.

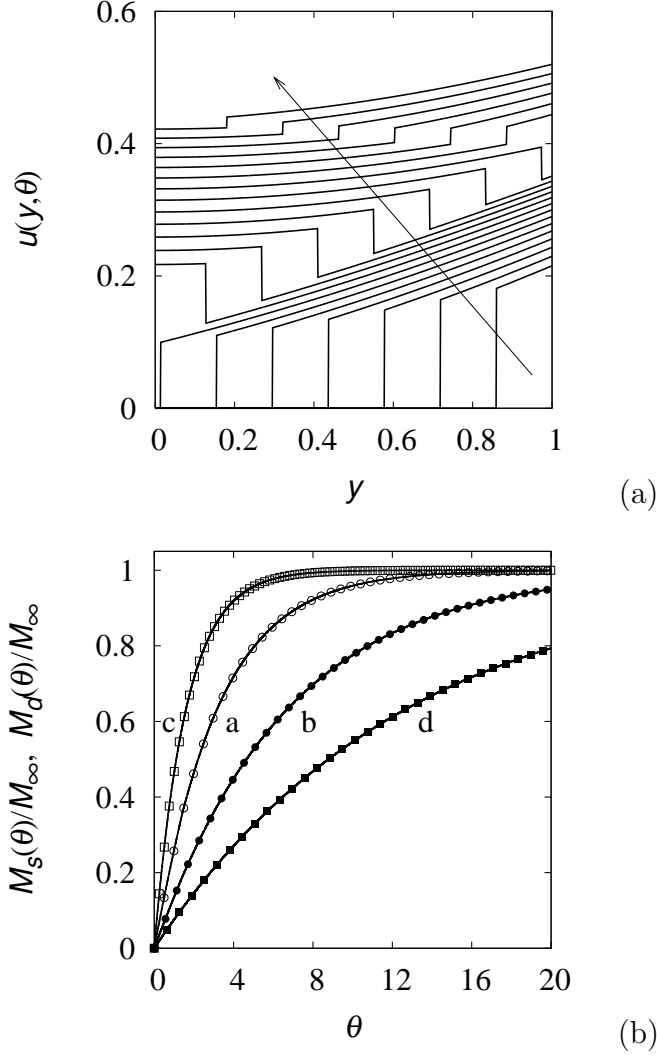


Figure 5: Panel (a): Concentration profiles in a sorption experiment at $\tau_c = 0.5$, imposing the boundary conditions (34) with $\gamma = 0.6$, sampled at $\Delta\theta = 0.1$ (time increases in the direction of the arrow). Panel (b): Sorption curves $M_s(\theta)/M_\infty$ from eq. (35) (solid lines) for different values of τ_c and γ . Symbols are the corresponding desorption curves $M_d(\theta)/M_\infty$. Line (a) and (\circ): $\tau_c = 0.5$, $\gamma = 0.6$, line (b) and (\bullet): $\tau_c = 0.5$, $\gamma = 0.8$, line (c) and (\square): $\tau_c = 0.1$ and $\gamma = 0.6$, line (d) and (\blacksquare): $\tau_c = 0.1$ and $\gamma = 0.95$.

(20) defined on an interval \mathcal{D} . So far no assumption is made for \mathcal{D} , be it $(-\infty, \infty)$ or $[0, L]$. Multiplying the first equation (20) by p^+ , the second by p^- , summing the resulting equations, and integrating over \mathcal{D} provides the

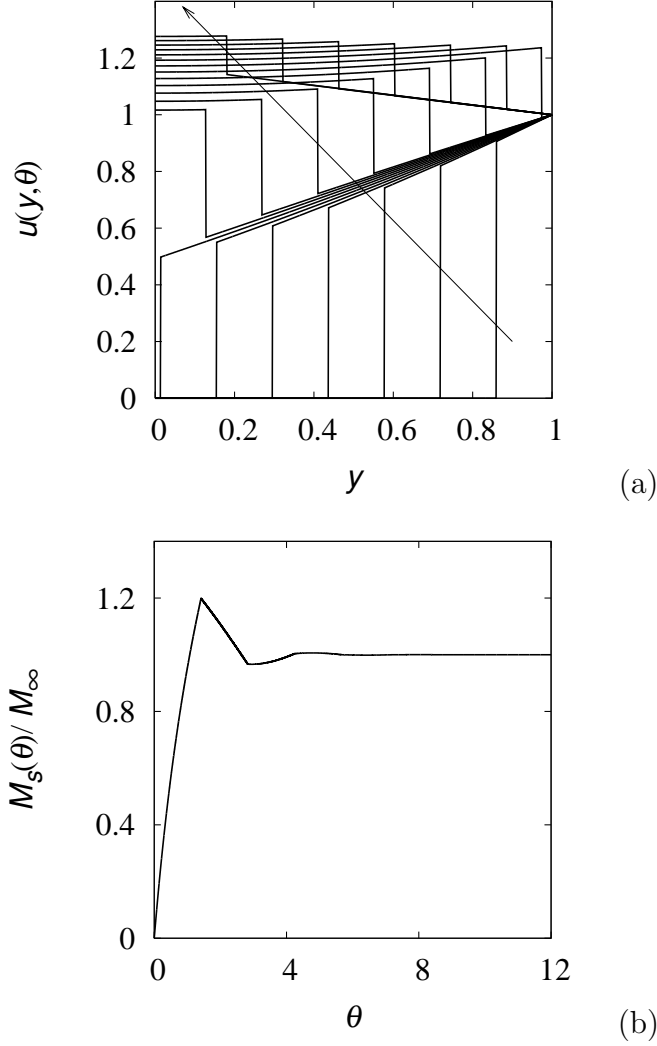


Figure 6: Panel (a): Concentration profiles $u(y, \theta)$ in a sorption experiment imposing the boundary condition (36), $\tau_c = 0.5$. The curves are sampled at $\Delta\theta = 0.1$ (time increases in the direction of the arrow). Panel (b) Sorption curve $M_s(\theta)/M_\infty$ associated with the data depicted in panel (a).

expression

$$\begin{aligned}
 \frac{1}{2} \frac{d}{dt} \int_{\mathcal{D}} [(p^+)^2 + (p^-)^2] dx &= -\frac{c}{2} \int_{\mathcal{D}} \frac{\partial}{\partial x} [(p^+)^2 - (p^-)^2] dx \\
 &\quad - a \int_{\mathcal{D}} (p^+ - p^-)^2 dx .
 \end{aligned} \tag{38}$$

Let us express the quantities entering eq. (38) in terms of concentration and flux:

$$\begin{aligned}(p^+)^2 + (p^-)^2 &= \frac{1}{2} (p^+ + p^-)^2 + \frac{1}{2} (p^+ - p^-)^2 = \frac{1}{2} p^2 + \frac{1}{2c^2} J^2 \\ (p^+)^2 - (p^-)^2 &= (p^+ + p^-)(p^+ - p^-) = \frac{1}{c} p J .\end{aligned}\quad (39)$$

Two cases can be considered: either $\mathcal{D} = (-\infty, \infty)$ or $\mathcal{D} = [0, L]$.

On the real line, i.e. $\mathcal{D} = (-\infty, \infty)$, regularity conditions at infinity apply, and consequently the first integral at the r.h.s. of eq. (38) vanishes identically.

Letting $\|f\|_{L^2}^2(t) = \int_{\mathcal{D}} f^2(x, t) dx$ be the square L^2 -norm of a square-summable real-valued function $f(x, t)$ in \mathcal{D} , eq. (38) can be rewritten as:

$$\frac{d}{dt} \left(\|p\|_{L^2}^2(t) + \frac{1}{c^2} \|J\|_{L^2}^2(t) \right) = -\frac{2}{k} \|J\|_{L^2}^2(t) , \quad (40)$$

which expresses the dissipation properties of hyperbolic “diffusion” i.e. of the stochastic motion driven by pure dichotomous Poisson noise. In the limit $c \rightarrow \infty$, $J \rightarrow -k\partial p/\partial x$, eq. (40) reduces to the classical norm condition, $d\|p\|_{L^2}^2(t)/dt = -2k \|\partial p/\partial x\|_{L^2}^2(t)$, characterizing Fickian diffusion.

Next, consider a bounded domain $\mathcal{D} = [0, L]$ and let us suppose (as in the previous Sections) that at $x = 0$ zero-flux conditions are enforced. Eq. (38) now becomes

$$\frac{d}{dt} \left(\|p\|_{L^2}^2(t) + \frac{1}{c^2} \|J\|_{L^2}^2(t) \right) = -I_{\text{boundary}} - \frac{2}{k} \|J\|_{L^2}^2(t) , \quad (41)$$

where the boundary term I_{boundary} equals

$$\begin{aligned}I_{\text{boundary}} &= 2p(L, t) J(L, t) \\ &= 2c [p^+(L, t) + p^-(L, t)] [p^+(L, t) - p^-(L, t)] .\end{aligned}\quad (42)$$

Irreversibility, or equivalently the dissipative nature of the process, imply that the r.h.s. of eq. (41) should be strictly non-positive. Consider at $x = L$ homogeneous boundary conditions for the partial waves, such as in eq. (30). A sufficient (not necessary) condition is that the constant γ entering eq. (30) satisfies the inequality $\gamma \leq 1$, which is the condition obtained in Section 4 from positivity arguments.

6 Biased hyperbolic transport and boundary-layer polarization

In this Section we extend the analysis of boundary conditions in hyperbolic models to biased transport. Specifically, let $v(x)$ be a deterministic velocity field, and consider the stochastic dynamics:

$$dx(t) = v(x(t))dt + c(-1)^{\chi(t)} dt , \quad (43)$$

defined in the interval $x \in [0, L]$. The PPM that specifies the statistical properties of eq. (43) is expressed by the system of two balance equations

$$\begin{aligned}\frac{\partial p^+(x, t)}{\partial t} &= -\frac{\partial [v(x) p^+(x, t)]}{\partial x} - c \frac{\partial p^+(x, t)}{\partial x} - a p^+(x, t) + a p^-(x, t) \\ \frac{\partial p^-(x, t)}{\partial t} &= -\frac{\partial [v(x) p^-(x, t)]}{\partial x} + c \frac{\partial p^-(x, t)}{\partial x} + a p^+(x, t) - a p^-(x, t)\end{aligned}\quad (44)$$

where $p^\pm(x, t)$ are defined as in eq. (19).

Let us study the evolution of the overall concentration field $p(x, t)$ and of its partial waves in a closed system, supposing that $v(0) = 0$, while $v(L) > 0$. For instance, consider the simple model for the biasing field

$$v(x) = -V_L \sin\left(\frac{3\pi x}{2L}\right), \quad (45)$$

where $V_L > 0$. Since $v(L) > 0$, there is a nonvanishing, outwardly directed, convective flux at $x = L$. The closedness of the system implies that the net flux vanishes at $x = 0, L$. Since $v(0) = 0$, reflecting boundary conditions for the partial waves apply at $x = 0$, i.e. eq. (24), as in the previous examples.

More interesting is the analysis at $x = L$, where the vanishing of the net flux dictates

$$v(L) p(L, t) + J(L, t) = 0, \quad (46)$$

where $J(L, t)$ is the “diffusive” (non-Fickian) flux associated with the stochastic Poisson perturbation in eq. (43). In terms of partial waves this implies

$$v(L) [p^+(L, t) + p^-(L, t)] + c [p^+(L, t) - p^-(L, t)] = 0, \quad (47)$$

meaning that the “diffusive” flux, proportional to $p^+ - p^-$, should counterbalance the convective one, proportional to $p^+ + p^-$, creating a concentration boundary layer (*polarization*) near $x = L$. Since $v(L) = V_L$, the latter condition becomes

$$(c - V_L) p^-(L, t) = (c + V_L) p^+(L, t). \quad (48)$$

Eq. (48) applies, without returning negative values of $p^-(L, t)$, if the condition

$$c > V_L, \quad (49)$$

is fulfilled. The case $c < V_L$ provides negative values of $p^-(L, t)$, while if $c \rightarrow V_L$ the backward wave at the boundary diverges, $p^-(L, t) \rightarrow \infty$.

Therefore, boundary layer polarization can be properly defined in hyperbolic transport models if eq. (49) is fulfilled, i.e., the intensity of the

stochastic velocity perturbation, namely c , is strictly greater than the velocity V_L at the boundary. In this case, the proper boundary condition for the partial waves at $x = L$ is homogeneous and equal to

$$p^-(L, t) = \Gamma p^+(L, t), \quad \Gamma = \frac{c + V_L}{c - V_L}, \quad 1 \leq \Gamma < \infty. \quad (50)$$

Let us consider a numerical example which highlights better the meaning and the nature of condition (49). We assume for $v(x)$ the expression (45) and let $L = 1$. Figure 7 (panel a) depicts the behavior of the steady-state distribution $p_s(x) = p_s^+(x) + p_s^-(x)$ associated with eq. (44), obtained from the asymptotic solution of PPM eq. (44) for $t \rightarrow \infty$. The values of the parameters are $\tau_c = 0.5$, and $k = 1$, and we set $V_L = \nu c$ with $\nu < 1$. The steady-state distributions refer to several values of $\nu = V_L/c$. Given τ_c and k , the parameters c , a specifying the stochastic perturbation are determined by the relations $a = 1/2\tau_c$, $c = \sqrt{2ka} = \sqrt{k/\tau_c}$. The numerical results of the PPM eq. (44), represented by solid lines in figure 7, are compared with stochastic simulations by considering an ensemble of $N = 2 \times 10^5$ particles following the stochastic micro-dynamics (43). The initial positions of the particles are uniformly distributed in the interval $[0, 1]$. Reflective boundary conditions are assumed at $x = 0$ and $x = 1$. Whenever a collision at the boundaries $x = 0$ or $x = 1$ occurs, say at $t = t^*$, the stochastic process is updated. This means that at $t = t^*$, the factor $(-1)^{\chi(t)}$ changes sign, namely, $(-1)^{\chi(t^*_+)} = -(-1)^{\chi(t^*_-)}$, where $t^*_\pm = \lim_{\varepsilon \rightarrow 0} t^* \pm \varepsilon$. As can be observed the agreement between PPM and stochastic simulations is excellent. The hyperbolic transport model provides the occurrence of a polarization layer near $x = 1$, that becomes more pronounced as ν tends to 1.

The maximum of the steady-state distribution occurs at some intermediate abscissa x^* , corresponding to the stable node of the forward wave, i.e., $v(x^*) + c = 0$, and not at $x = 0$ which represents the stable equilibrium point of the deterministic dynamics.

Keeping track of the values of $(-1)^{\chi(t)}$, it is possible to extract from stochastic simulation data the distribution of the two-partial waves $p_s^+(x)$ and $p_s^-(x)$, using their definitions (18) as joint probabilities with respect to the dichotomous outcome of $(-1)^{\chi(t)}$. The comparison between PPM and stochastic simulations for the partial waves is depicted in figure 7 (panel b), revealing also in this case an excellent agreement, limited solely by the relatively small size, $N = 2 \times 10^5$, of the particle ensemble considered in the statistics.

The comparison of PPM simulation and stochastic data reveals an interesting property of the condition (49). In point of fact, there are neither physical limitations nor fundamental consistency constraints in simulating an ensemble of particles moving according to eq. (43) beyond the limit imposed by eq. (49), i.e., for $V_L > c$, imposing reflective conditions at the boundaries.

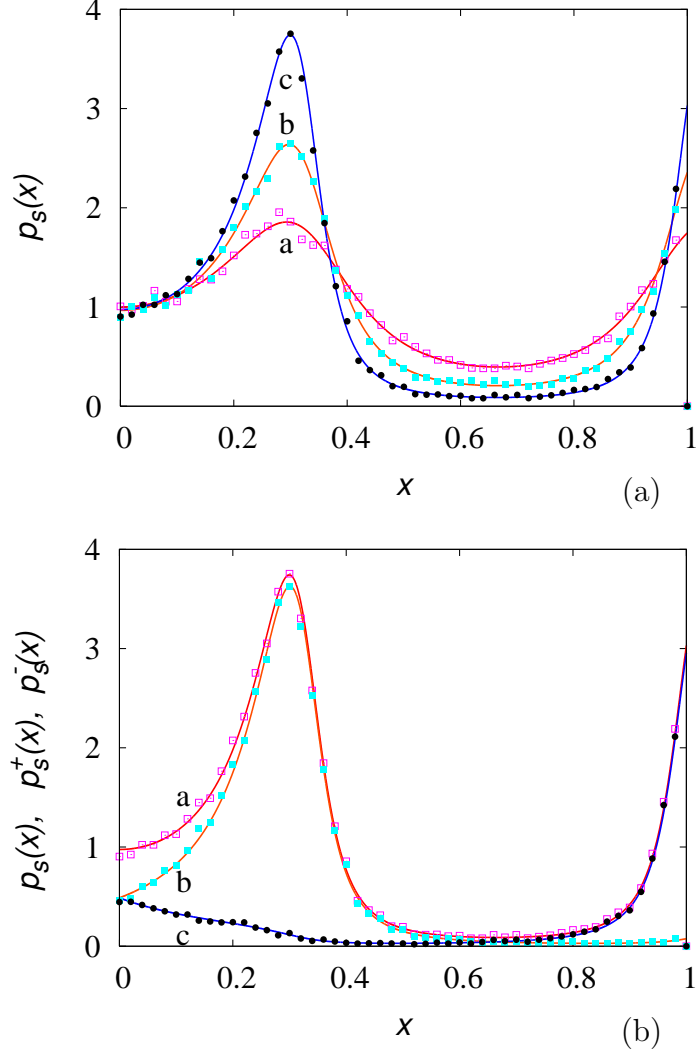


Figure 7: Panel (a): Unique-steady state probability density function $p_s(x)$ vs x for $\tau_c = 0.5$, $k = 1$, at different values of ν . Solid lines are the results of the partial wave model (44), symbols represent stochastic simulations for an ensemble of particles evolving according to eq. (43). Line (a) and (□): $\nu = 0.8$, line (b) and (■): $\nu = 0.9$, line (c) and (●): $\nu = 0.95$. Panel (b): Steady-state probability density function $p_s(x)$ vs x (line (a) and symbols (□)), and associated steady-state partial waves $p_s^+(x)$ (line (b) and symbols (■)) and $p_s^-(x)$ (line (c) and symbols (●)) for $\tau_c = 0.5$, $k = 1$, $\nu = 0.95$.

While eq. (28) stems from physical principles (non-negativity of probabilities, or equivalently dissipativity as developed in Section 5), this is not the case of eq. (49). The origin of the critical condition $c = V_L$ is not

of basic physical nature, but possesses a dynamic explanation. Namely, it is essentially a bifurcation point of the partial-wave model, associated with the ergodicity-breaking [28], corresponding to the birth of multiple invariant measures. For $c > V_L$ there is a unique invariant stationary probability density function $p_s(x)$. Conversely, for $c < V_L$, the stochastic dynamics (43) is characterized by the occurrence of two disjoint invariant intervals $I_1 = [0, x^*]$ and $I_2 = [x^{**}, 1]$, where x^* is the first root of the equation $v(x^*) + c = 0$ and x^{**} the root of $v(x^{**}) - c = 0$ (see figure 8). Consequently there exist two distinct steady-state distributions $p_{s,h}(x)$, $h = 1, 2$ localized within the two intervals I_h .

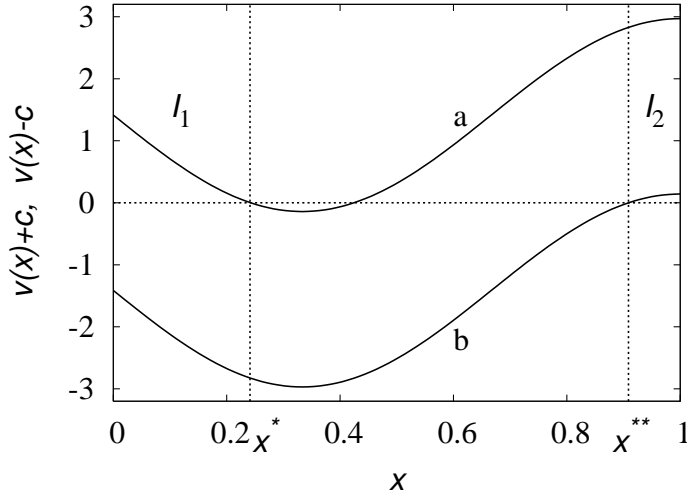


Figure 8: $v(x) + c$ (line a) and $v(x) - c$ (line b) for $\nu = V_L/c = 1.1$. Dotted vertical lines mark the two characteristic points x^* , x^{**} representing the endpoints of the two invariant intervals $I_1 = [0, x^*]$ and $I_2 = [x^{**}, 1]$.

This phenomenon can be easily verified via stochastic simulations starting from different initial distributions, as depicted in figure 9 panels (a)-(c) for $\nu = 1.1$. In this case, $x^* \simeq 0.2412$ and $x^{**} \simeq 0.9087$.

Panel (b) depicts the steady-state distributions obtained by considering an initial particle ensemble uniformly distributed in $[0, 0.3] \subset I_1$, returning an absolutely continuous invariant distribution localized near x^* . Conversely for an initial ensemble contained in I_2 (depicted in panel (c)), an impulsive invariant distribution $p_{s,2}(x)$ localized at $x = 1$ is obtained. The simulation starting from a uniform distribution throughout $[0, 1]$, depicted in panel (a), provides a mixed steady-state distribution $p_s(x) = \alpha p_{s,1}(x) + (1 - \alpha)p_{s,2}(x)$, $0 < \alpha < 1$, that is a convex combination of the two $p_{s,h}(x)$.

Although a thorough analysis of ergodicity-breaking bifurcations is developed elsewhere [28], since it goes beyond the analysis of boundary condi-

tions representing the focus of the present work, it is interesting to analyze the boundary conditions that apply to the partial-wave model for $V_L > c$ in order to reproduce the results obtained from stochastic simulations. Also in the analysis of this issue, the answer stems from a wave-oriented interpretation of hyperbolic transport.

With the aid of figure 8, a wave-dynamic interpretation of the bifurcation occurring at $V_L = c$ can be achieved. From their definition and dynamics (44), the partial probability $p^+(x, t)$ and $p^-(x, t)$ corresponds to waves that propagate with velocities $v^+(x) = v(x) + c$ and $v^-(x) = v(x) - c$, respectively. For $V_L < c$, in the neighbourhood of $x = 1$, $v^+(x) > 0$ and $v^-(x) < 0$, thus representing properly a progressive and a regressive wave, respectively.

In this case, the balance at $x = 1$ of the overall flux permits to derive the condition (50) for the “reflection coefficient” Γ , providing a regressive wave that balances exactly the outward probability flux carried out by the progressive wave.

The bifurcation point $V_L = c$ marks the transition for $p^-(x, t)$ from a regressive wave ($v^-(x) < 0$) to a progressive wave ($v^-(x) > 0$), near $x = 1$. In the latter case, impermeability at $x = 1$ cannot be longer enforced as relation connecting $p^-(1, t)$ to $p^+(1, t)$, since $p^-(1, t)$ would be negative. It is rather a consequence of the implicit assumption assumed in stochastic simulations of an arbitrarily large (and in principle infinite) potential barrier at $x = 1$ originating the reflective condition for eq. (43) at $x = 1$.

In order to derive the proper boundary conditions it is therefore convenient to consider the restriction of eq. (44) to each of the two invariant intervals I_h , $h = 1, 2$. Consider eq. (44) restricted to $x \in I_1 = [0, x^*]$. At $x = 0$, the usual reflective condition applies. At the other endpoint $x = x^*$ of the invariant interval, zero net-flux condition should be enforced, namely,

$$[v(x^*) + c] p^+(x^*, t) + [v(x^*) - c] p^-(x^*, t) = 0. \quad (51)$$

Since $v(x^*) + c = 0$, eq. (51) implies

$$p^-(x^*, t) = 0, \quad (52)$$

which indicates that the regressive wave should vanish at x^* . Figure 10 depicts the comparison of the PPM (43) restricted to I_1 (solid lines) equipped with the boundary condition (52) and the results of stochastic simulation for $p_{s,1}(x)$ and its partial components $p_{s,1}^+(x)$, $p_{s,1}^-(x)$ at $\nu = 1.1$. An ensemble of $N = 10^6$ particles has been used in the stochastic simulations. As expected from eq. (52), stochastic simulation data confirm the vanishing value of the regressive wave at x^* .

Consider $x \in I_s = [x^{**}, 1]$. Similarly to eq. (52) one derives

$$p^+(x^{**}, t) = 0, \quad (53)$$

since $v^-(x^{**}) = v(x^{**}) - c = 0$.

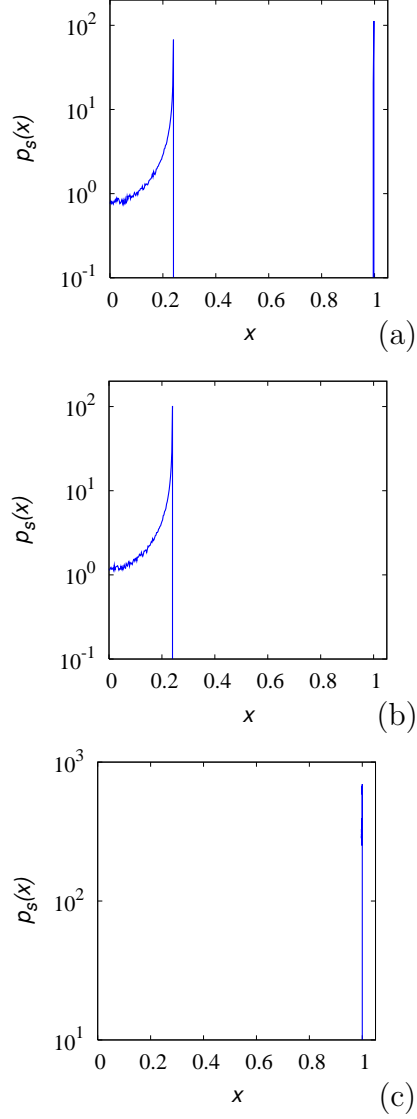


Figure 9: Steady-state distributions for $\tau_c = 0.5$, $k = 1$, $\nu = 1.1$ (i.e., violating condition (49)) obtained from stochastic simulation of eq. (43) for different distributions of initial conditions. Panel (a): uniform distribution in $[0, 1]$, panel (b): uniform distribution in $[0, 0.3]$, panel (c): uniform distribution in $[0.92, 1]$.

As stated above, the boundary condition at $x = 1$ does not derive from a balance between partial waves (as both are progressive waves in the neighbourhood of $x = 1$), but is a consequence of the assumption of an infinite potential barrier preventing particles to escape from the boundary in

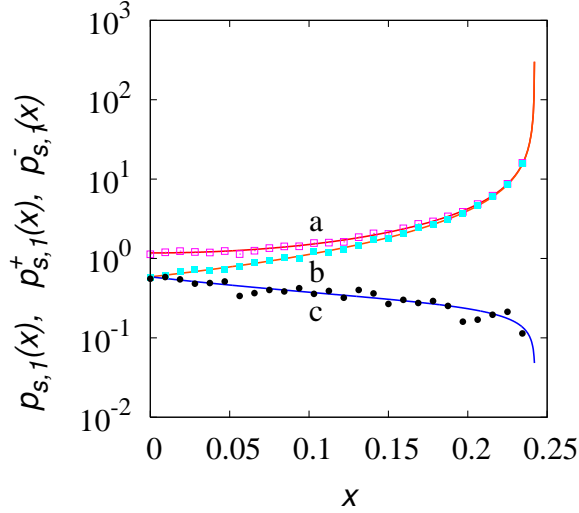


Figure 10: Steady-state distributions $p_{s,1}(x)$, $p_{s,1}^{\pm}(x)$ restricted to I_1 for $\tau_c = 0.5$, $k = 1$, $\nu = 1$, 1 . Solid lines represent the result of the partial wave model enforcing condition (52), symbols correspond the stochastic simulation data. Line (a) and (\square): $p_{s,1}(x)$, line (b) and (\blacksquare): $p_{s,1}^+(x)$, line (c) and (\bullet): $p_{s,1}^-(x)$.

stochastic simulations.

This observation implies that the partial waves possess an absolutely continuous component $\tilde{p}^{\pm}(x, t)$ and an impulsive part localized at $x = 1$,

$$p^{\pm}(x, t) = \tilde{p}^{\pm}(x, t) + \pi^{\pm}(t) \delta(x - 1), \quad (54)$$

where the values of $\pi^{\pm}(t)$ stem from probability conservation. Asymptotically, a steady impulsive distribution is obtained, $p_{s,2}(x) = \delta(x - 1)$, as physically expected since $v^{\pm}(x) \geq 0$ uniformly in I_2 , as can be observed in figure 9 in panels (a) and (c).

7 Concluding remarks

In this article we have analyzed in detail the structure and the admissibility of different forms of boundary conditions for one-dimensional hyperbolic transport problems on the interval, both in the absence and in the presence of a deterministic biasing field.

We stress again that the concept of a *hyperbolic transport model* should be regarded as a system of balance equations, obtained from the underlying micro-dynamics expressed in the form of a stochastic differential equation driven by dichotomous noise and its generalizations. In this framework, hyperbolic transport theory is the wave-like counterpart of the classical trans-

port approach in which the stochastic micro-dynamics is controlled by a Langevin equation driven by Wiener processes (Brownian motion).

In this concluding Section we would like to pinpoint some general observations emerging from the present work that can be used to develop a self-consistent theory of hyperbolic transport, not limited to one-dimensional problems, and that be will thoroughly elaborated elsewhere:

1. The primitive observables in hyperbolic transport theory are the partial probabilities (in an one-dimensional model with a single velocity c , these are just the partial probability waves $p^+(x, t)$ and $p^-(x, t)$). The existence of these observables derives intrinsically from the dichotomous nature of the underlying stochastic fluctuations at microscale.
2. The overall concentration $p(x, t)$ as well as its flux $J(x, t)$ should be regarded in the theory as *derived* quantities, although they represent the physical observables commonly measured in transport experiments. This result is somehow similar to the setting of non-relativistic quantum mechanics where the wave function is the primitive quantity of the theory, while the probabilistic description of a quantum system is associated to the square of its modulus (Born condition).
3. Boundary conditions in the hyperbolic theory are written as relations connecting forward and backward probability waves, corresponding to transmission/reflection conditions for the partial probability waves. The set of admissible boundary conditions is dictated by the positivity of partial probability functions, and can be derived alternatively by invoking the dissipative nature of the dynamics (Section 5).
4. There is a close connection between the functional relation describing dissipation in hyperbolic “diffusion” eq. (40) and the approach followed in *Extended Irreversible Thermodynamics* [10, 11, 12], where the entropy function depends also on the flux and not only on the concentration. The observation that the basic primitive quantities in hyperbolic transport theory are the partial waves suggests that the thermodynamic formalism developed in the theory of *Extended Irreversible Thermodynamics* could be reformulated and/or generalized in order to include the primitive variables of hyperbolic transport theory (namely the partial probabilities) as the system variables with respect to which the state functions (such as entropy) are expressed. This effort would permit to resolve the problem of the absence of any probabilistic meaning of the Cattaneo equation (which is the prototype of generalized transport equation with memory in extended thermodynamics) in space dimensions greater than one.

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